Quantum chaos and fluctuations in isolated nuclear spin systems

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Using numerical simulations we investigate dynamical quantum chaos in isolated nuclear spin systems. We determine the structure of quantum states, investigate the validity of the Curie law for magnetic susceptibility and find the spectrum of magnetic noise. The spectrum is the same for positive and negative temperatures. The study is motivated by recent interest in condensed-matter experiments for searches of fundamental parity- and time-reversal-invariance violations. In these experiments nuclear spins are cooled down to microkelvin temperatures and are completely decoupled from their surroundings. A limitation on statistical sensitivity of the experiments arises from the magnetic noise.

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Introduction. Much of the present knowledge about violations of the fundamental symmetries P (invariance under spatial inversion) and T (invariance with respect to time reversal) comes from experiments measuring P- and T-violating permanent electric dipole moments (EDM's) of atoms, molecules and the neutron, see, for example, Ref. [1]. Most EDM experiments measure precession of the angular momentum of the system in an applied electric field analogous to the Larmor precession in an applied magnetic field.

In addition to such precession experiments, there are EDM searches of another kind [2, 3], which have drawn recent renewed attention [4, 5, 6, 7, 8, 9, 10]. The idea of these experiments is the following. Suppose that we have some condensed matter sample with uncompensated spins. If an electric field is applied to the sample, it interacts with the associated (P- and T-violating) EDM's, leading to a slight orientation of the spins in the direction of the electric field. This orientation, in turn, is measured by measuring the induced magnetization of the sample. In this work we concentrate on effects related to nuclear spins in insulators with fully compensated electron spins [7, 9]. The EDM energy shift under discussion is about $10^{-24} - 10^{-28}$eV per nuclear spin. For comparison a similar shift is created by a magnetic field $B \sim 10^{-16} - 10^{-20}$T. So the effect is tiny and limitation to statistical sensitivity comes from the fact that the number of spins, $n \sim 10^{23}$, inspite of being large is still finite [9]. Basically the limitation comes from a kind of magnetic shot noise. In the EDM experiments the nuclear spins must be cooled down at least to 100μK and optimally down to $10 - 100$nK. At low temperatures the spins are completely decoupled from the crystal lattice, so there is no contact with any heat bath. This motivates the problem considered in the present work: magnetic noise of an isolated nuclear spin system. An important point is that the total spin of all nuclei is not conserved because magnetic dipole-dipole interaction depends on relative orientation of nuclei.

Concerning previous work we first of all refer to the 1959 paper by Hebel and Slichter [11] where the physical meaning of temperature for an isolated spin system has been discussed. This work assumed the validity of a statistical approach for the isolated quantum system. This is what nowadays is called dynamical quantum chaos. The problem of the onset of dynamical quantum chaos has been addressed much later. Level statistics in a spin system with a mobile fermion has been investigated by Montambaux et al in 1993 [12]. The criterion for onset of quantum chaos in spin glass shards with Heisenberg interaction in a random external magnetic field has been derived by Georgeot and Shepelyansky in 1998 [13]. There have been also numerical studies of level statistics in XYZ spin chains with and without a random magnetic field that demonstrated non-Wigner behavior in the absence of magnetic field [14].

Model. To be specific, we consider the Lead Titanate ferroelectric suggested for EDM experiments in Refs. [7, 15]. The interaction between the nuclei is governed by the magnetic dipole-dipole interaction, the strength of which falls off as $1/r^3$, $J_{\alpha\beta} = \gamma^2(\delta_{\alpha\beta} - 3n_\alpha n_\beta)/r^3$. Therefore in the paramagnetic phase that we are interested in it is sufficient to only include nearest-neighbour interactions. The dominating contribution to the dipole-dipole interaction is from the ^{207}Pb isotope which has a natural abundance of 22.1% and are distributed randomly through the lattice. As a consequence the interaction is anisotropic and random in strength. Therefore we adopt a model Hamiltonian of the form,

$$H = \sum_{\langle kl \rangle} \sum_{\alpha\beta} J^{kl}_{\alpha\beta} S_{k\alpha} S_{l\beta} - B \sum_k S_{kz} \tag{1}$$

where $J^{kl}_{\alpha\beta}$ is the interaction between spins $S = 1/2$ on nearest sites k and l on a lattice and B is a uniform external magnetic field. The interaction $J^{kl}_{\alpha\beta}$ is represented by random numbers uniformly distributed between [-J,J]. The typical value of J is $J \sim 10^{-12}eV \sim 10nK$ [7]. For the sake of simplicity unless otherwise is stated we take a usual square lattice with periodic boundary conditions. We will consider the case when the tensor $J_{\alpha\beta}$ has only diagonal components, $\alpha = \beta$, and the case when it has both diagonal and off diagonal components. The latter

case corresponds to real dipole-dipole interaction. We diagonalize the Hamiltonian (1) numerically exactly and we need to know all eigenstates and all eigenenergies, so the size of the matrix is 2^n where n is the number of spins. Therefore, practically we are able to consider only relatively small clusters with n=8,10,12. For a sufficiently large lattice all results are expected to be self-averaged. However, we consider relatively small clusters, therefore, to improve statistics we average results over 100 random realizations of $J^{kl}_{\alpha\beta}$. We do not consider an odd number of spins to avoid Kramers degeneracy of spectra that cannot be important in the thermodynamic limit.

Structure of chaotic eigenstates. Small magnetic field. At zero magnetic field the Hamiltonian (1) is invariant under time reversal (T-reversal). This implies that the eigenfunctions of the Hamiltonian are split in two sectors of different time-parity: in the first sector they stay the same (+) and in the second one they change sign (-) under the action of all the spins being flipped. Therefore the expectation value of magnetization that is a T-odd operator vanishes for any state, $\langle i|S_z|i\rangle = 0$. Because of random J, chaos is established in each of these two sectors but the sectors do not interact. Anderson localization of single particle states in a random potential is a well known effect. In principle a spatial localization of many-body quantum states is also possible, while it is known that interaction tends to destroy Anderson localization [16]. Spatial localization would imply Poisson level statistics (the distribution of level spacing between closest levels), $P_P(s) = \exp(-s)$, within a sector with a given T-parity in a sufficiently large system. On the other hand the Wigner-Dyson distribution $P_{WD}(s) = \frac{\pi s}{2}\exp(-\pi s^2/4)$ within a given sector indicates a full chaotization including delocalization. To characterise to what degree the statistics of levels are Wigner or Poisson, following Ref. [13] we use the parameter $\eta = \int_0^{s_0}[P(s)-P_{WD}(s)]\,\mathrm{ds}/\int_0^{s_0}[P_P(s)-P_{WD}(s)]\,\mathrm{ds}$, where $P(s)$ are the statistics measured in numerical simulations and $s_0 = 0.4729$ is the intersection point of $P_P(s)$ and P_{WD}. Localization effects are always enhanced in the 1D case. Therefore to investigate the localization scenario we studied level statistics within a given T-parity sector for the Hamiltonian (1) on a 1D ring at $B = 0$. We found that $\eta = 0.18, 0.051, 0.035$ for n=8, 10 and 12 respectively. So, we do not observe any deviation from the Wigner-Dyson distribution within the accessable system size. Thus, we come to a conclusion that there is no spatial localization of quantum states in the random spin system. This conclusion is similar to that of Ref. [16] for mobile interacting particles.

The combined statistics of levels including both sectors at $B = 0$ is given by the sum of two Wigner distributions, giving an intermediate statistics with $\eta \approx 0.5$, see Fig.1. A very small critical magnetic field is needed to mix the sectors and hence to lead to a single Wigner distribution. The critical magnetic field B_{c1} is given by the condition that mixing of two nearest states from different sectors is of the order of unity

$$\frac{B_{c1}\langle j_-|S_z|i_+\rangle}{\Delta E} \sim 1\ . \tag{2}$$

Here $|i_+\rangle$ and $|j_-\rangle$ are opposite T-parity eigenstates of the Hamiltonian (1) at zero magnetic field. The level spacing is roughly equal to $\Delta E \sim J\sqrt{n}/2^n$, where the factor $\sqrt{n}$ comes from the total width of the spectrum that is discussed below. To estimate a typical mixing matrix element in (2) we use the sum rule $\sum_j \langle i_+|S_z|j_-\rangle\langle j_-|S_z|i_+\rangle = \langle i_+|S_z^2|i_+\rangle = \langle i_+|\sum_m S^2_{zm} + \sum_{m\neq k} S_{z,m}S_{z,k}|i_+\rangle \approx \frac{n}{4}$. Here we take into account that $\sum_m S^2_{zm} = n/4$ and that $\sum_{m\neq k} \to 0$ because it is incoherent. There are 2^{n-1} terms in $\sum_j$. Therefore the typical mixing matrix element is $|\langle j_-|S_z|i_+\rangle| \sim \sqrt{n/2^{n+1}}$. Hence substitution in (2) gives the following estimate

$$B_{c1} \sim \frac{J}{\sqrt{2^{n-1}}}\ . \tag{3}$$

To confirm this analytical estimate we present in Fig. 1 the value of η calculated numerically for different magnetic fields. As we already mentioned at zero magnetic field $\eta \approx 0.5$ indicating intermediate statistics, and at large field $\eta \to 0$ indicating the Wigner-Dyson distribution. Taking the value $\eta = 0.25$ as a crossover point we find $B_c(n+2)/B_c(n) \approx 0.5$ in good agreement with Eq. (3). Thus the value of the first critical field B_{c1} drops

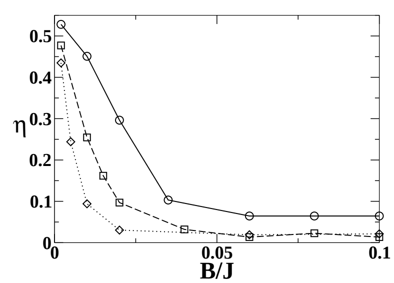

FIG. 1: η as a function of the magnetic field: solid line $n = 8$, dashed line $n = 10$, dotted line $n = 12$.

down exponentially with number of spins. For the EDM experiment we are interested in large systems, $n \sim 10^{23}$. So in this situation $B_{c1} = 0$. Note that Eq. (3) is not a criterion for onset of quantum chaos. This is the criterion for a crossover from a two component chaotic distribution to a single component chaotic distribution or in other words the criterion for destruction of T-parity. This is quite similar to the dynamical enhancement of spatial parity violation in nuclei [17].

"Strong" magnetic field. In a strong uniform magnetic field stationary states can be classified by total spin projection on the direction of the field, S_z. This regime is realized when the level spacing B is larger than the matrix element between directly coupled states that is of the order of J, see e. g. [13]. Thus the

crossover to the strong magnetic field regime happens when $B > B_{c2} \sim J \sim 10^{-7}$T (the numerical value corresponds to Lead Titanate). In the strong field regime due to random exchange interaction J the states with a given value of S_z are completely mixed up and their energies, according to our calculations, are spread within the band of width $\approx 0.3\sqrt{n}J$. Note that "conservation" of total S_z does not mean that the z-projection of a particular spin is conserved. Every particular spin fluctuates very strongly. Thus the energy spectrum consists of successive bands, separated by B, the width of the band is about $\sim 0.3\sqrt{n}J$. There is the Wigner-Dyson statistics of levels within a given band. When $n \gg (B/J)^2$ the bands overlap and this leads to Poisson level statistics.

The above discussion of the structure of quantum states is valid in the case when the tensor $J_{\alpha\beta}$ in Hamiltonian (1) has both diagonal and off diagonal components. In the case without off diagonal components there is a hidden integral of motion. The Hamiltonian acting on a basis state with a particular S_z only changes S_z by $0, \pm 2$, therefore $S_z(mod 2)$ is conserved. This means that eigenfunctions can be expanded in a basis with either even or odd S_z components. Then the above considerations are valid separately for S_z-even and for S_z-odd sectors. It is possible that the non-Wigner behaviour observed in Ref.[14] is due to the hidden integral of motion.

Temperature, average energy and magnetic susceptibility. The textbook analysis of a spin system based on assuming the validity of the Boltzmann distribution gives the following well known relations [18]

$$\chi = \frac{n}{4T} \ , \quad E(T) = \overline{E}_i - \frac{1}{T}\left(\overline{E_i^2} - \overline{E}_i^2\right) \ . \tag{4}$$

Here χ is the magnetic susceptibility, $E(T)$ is the average energy at a given temperature (we set the Boltzmann constant equal to unity), $\overline{E}_i$ is the average energy of stationary states and $\overline{E_i^2}$ is the average energy squared. $\overline{E}_i$ and $\overline{E_i^2}$ are independent of temperature. First we want to check the validity of Eq.(4) for the isolated dynamical system (1). In this case Eq.(4) in essence defines an effective temperature, see the discussion in Ref.[11]. To give a precise meaning to Eq. (4) one has to consider many energy levels inside a bin around some given energy E. Then, according to (4) the susceptibility averaged over these levels is related to the energy E. We have checked numerically that for the Hamiltonian (1) $\overline{E}_i \approx 0$ and $\overline{E}_i^2 \approx 0.13J^2 n$. Hence, according to (4) $\chi \approx -1.94E/J^2$. This agrees well with results of numerical simulations with Hamiltonian (1) shown in Fig. 2a. This figure represents values of the level magnetization $\langle i|S_z|i\rangle$ calculated in the field $B = 0.1J$, averaged over bins of width $\Delta E = 0.5J$, and averaged over random $J_{\alpha\beta}$. Deviation from linear dependence at large positive and negative energy is the finite size effect: Eqs. (4) make sense only if the average energy is smaller than the "band width", $|E| \lesssim \sqrt{n}J$. It is instructive to consider instead

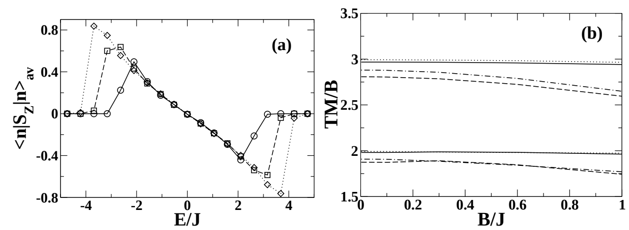

FIG. 2: **a**. Magnetization $\langle i|S_z|i\rangle$ averaged over energy bins $\Delta E = 0.5J$. The magnetic field $B = 0.1J$. Solid, dashed, and dotted lines correspond to clusters of size $n = 8, 10, 12$ respectively.
b. The value TM/B versus magnetic field for temperatures T=-3J (solid), -J (dashed), J (chain), 3J (dotted) and for cluster sizes $n = 8, 12$. The lower group of lines correspond to $n = 8$ and the upper group to $n = 12$.

of "rectangular binning" another way of sampling of energy levels. In particular the Boltzmann sampling defined as $M = \frac{1}{Z}\sum_i \langle i|S_z|i\rangle e^{-E_i/T}$, where $Z = \sum_i e^{-E_i/T}$. Let us stress that temperature here is just a parameter that samples a set of energy levels around the average energy that according to (4) is $E = -0.13nJ^2/T$. One can also check that the energy "window" around the average energy is $\delta E_{rms} = \sqrt{0.13n}J$. Note that the Boltzmann sampling does not imply a validity of the equilibrium Boltzmann distribution. For example we can use Boltzmann sampling in a system without interaction that never comes to an equilibrium. The value of TM/B calculated for different cluster sizes, temperatures (positive and negative) and magnetic fields and averaged over random $J_{\alpha\beta}$ is presented in Fig. 2b. For high temperatures, $|T| = 3J$, the results agree perfectly with the Curie law. Unexceptedly it agrees even at $B \lesssim B_{c1}$. For lower temperatures, $|T| \sim J$ the results deviate from Curie's law as one should expect. To complete the analysis we have also calculated the dispersion of the susceptibility and found that $\delta\chi = \sqrt{\langle\chi^2\rangle - \langle\chi\rangle^2} \sim 0.01\sqrt{n}/T$.

Magnetic susceptibility at nonzero frequency, fluctuations. There are no temporal fluctuations in an isolated system in a given quantum state. The fluctuations come from the fact that a large system cannot be prepared in a fixed quantum state. We always deal with a density matrix that represents a combination of a large number of quantum states around some average energy. In this section we consider only the Boltzmann sampling: the average energy is $E = -0.13nJ^2/T$ and the energy "window" is $\delta E_{rms} = \sqrt{0.13n}J$. In this case we can use the standard formula for the imaginary part of the magnetic susceptibility [18]

$$Im[\chi(\omega)] = \frac{\pi}{\hbar}\left(1 - e^{-\hbar\omega/T}\right)\frac{1}{Z}\sum_{i,j} e^{-E_i/T} \times |\langle i|S_z|j\rangle|^2 \delta(\omega + \omega_{ij}) \ . \tag{5}$$

Here $\omega_{ij} = (E_i - E_j)/\hbar$. First we perform direct numer-

ical simulations using this formula and quantum states generated by the Hamiltonian (1) at $B = 0.1J$ [19] For simplicity we consider the case when the tensor $J_{\alpha\beta}$ has only diagonal components. Results of simulations for cluster $n = 12$ are shown in Fig. 3. The plots for $n = 8, 10$ are very similar. The result unambiguously in-

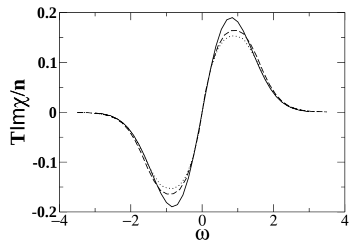

FIG. 3: $TIm\chi/n$ versus ω. Solid line, analytic. Numerical results for $n = 12$ at $T = 3$ and $T = 1$ are shown by dashed and dotted lines respectively.

dicates a Gaussian line shape, $Im[\chi] \approx \frac{n}{4T}\sqrt{\pi}\omega\tau e^{-\omega^2\tau^2}$, with $\tau \approx 0.85/J$. Assuming the Gaussian shape we can also calculate τ analytically using a method usually applied in NMR studies [20]. At a large temperature, $E_i \ll T$, and hence we can represent (5) as $Im[\chi(\omega)] = \frac{\pi\omega}{T}f(\omega)$ where the lineshape $f(\omega)$ is given by, $f(\omega) = \sum_{i,j} |\langle i|S_z|j\rangle|^2\delta(\omega + \omega_{ij})$. Let us calculate the second moment of $f(\omega)$, $M_2 = \int_{-\infty}^{\infty}\omega^2 f(\omega)\mathrm{d}\omega \Big/ \int_{-\infty}^{\infty} f(\omega)\mathrm{d}\omega$. Using the completeness relation and the spin decoupling $S_z^2 = \sum_{kl} S_{zk}S_{zl} \to 1/4\sum_{kl}\delta_{kl}$ (the indexes k, l enumerate sites) we find the denominator $\int_{-\infty}^{\infty} f(\omega)\mathrm{d}\omega = n2^{n-2}$. To calculate the numerator one represents it in terms of the commutator of spin with the Hamiltonian $\int_{-\infty}^{\infty}\omega^2 f(\omega)\mathrm{d}\omega = \sum_i\langle i|[H, S_z]^2|i\rangle$. Then calculating the commutator and using the spin decoupling we find

$$\int_{-\infty}^{\infty}\omega^2 f(\omega)\mathrm{d}\omega = \frac{2^n}{12}\sum_{\langle kl\rangle}\left[(J_{xx}^2 + J_{yy}^2 + J_{zz}^2)\right. \quad (6)$$
$$\left. -(J_{xx}J_{yy} + J_{yy}J_{zz} + J_{xx}J_{zz}) + 3(J_{xy}^2 + J_{zy}^2 + J_{xz}^2)\right] .$$

Here $\langle kl\rangle$ represents a pair of nearest sites. Since we have performed numerical simulations without off diagonal components of $J_{\alpha\beta}$, the right hand side of Eq. (6) is $n2^{n-1}J^2/3$ and hence $M_2 = 2/3J^2$. On the other hand for the Gaussian line shape $M_2 = 1/(2\tau^2)$. Hence the analytical calculation gives the $\tau = \sqrt{3/4}/J$. The Gaussian curve with this value of the relaxation time is shown in Fig. 3 by solid line. It is in good agreement with results of direct numerical simulations. The real part of the susceptibility can be easily found using Kramers-Kronig relations (see, for example, Ref. [18])

As soon as $Im[\chi]$ is known then using the fluctuation dissipation theorem one can find magnetic noise, i. e. the spectral density $(M^2)_\omega$ of the square of the deviation of the magnetization from its average value [9], $V^2(M^2)_\omega = \hbar\coth(\hbar\omega/2T)Im[\chi(\omega)]$. V is volume of the sample. A very interesting point is that at $\omega \ll T$ the noise is independent of temperature and moreover it is the same for positive and negative temperature. What is usually called the fluctuation dissipation theorem in the case of negative temperature becomes the fluctuation *radiation* theorem because $Im[\chi(\omega)]$ changes sign.

We have investigated the structure of chaotic quantum states in a spin lattice system with random interactions. We also checked the validity of the Curie law for magnetic susceptibility and find the spectrum of magnetic noise. The temperature independent noise limits the statistical sensitivity of experiments on parity and time-reversal-invariance violations.

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